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**PRODUCTION, MANIPULATION, AND APPLICATIONS OF ULTRACOLD POLAR MOLECULE**

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UNIVERSITY OF CONNECTICUT**

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Final Report**

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<b>14. ABSTRACT</b> The development of techniques for producing, trapping, and detecting ultracold polar molecules, including "assembly" of molecules from ultracold atoms and the direct cooling of both neutral and ionized molecules is the first theme of the project. The second is controlling the arising interactions using external electromagnetic fields, thus manipulating collisions and chemical reactions, and engineering molecule-based quantum information. The third is development of applications for ultracold polar molecules, such as technologies for high-resolution spectroscopy, engineering of highly-correlated interacting many-body systems. The outcome includes the first steps towards quantum-degenerate gases of polar molecules for a chemically diverse range of species, which have been trapped and cooled to the ultracold regime. Novel types of collisional and chemical-reactive behavior were observed, controlled, and understood theoretically. The first implementations of quantum information encoding and processing with polar molecule qubits were demonstrated, and improved schemes for large-scale systems are being devised. Novel features of many-body systems of strongly-interacting polar molecules were analyzed, and the details of molecular structure were measured and applied.						
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# Final Report, April 2015

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## Main accomplishments:

### Publications

Total: 130, of those: 8 Nature, 1 Science, 27 Phys. Rev. Lett.

### Milestones

The vast majority of milestones were reached and often surpassed, such as laser cooling, building a MOT, and buffer gas cooling, which was done for a larger variety of molecules and to a larger extent than originally proposed. The exception is the full cooling of  $\text{SrCl}^+$  to  $< 1$  mK, only part of this milestone was reached. For the theoretical projects, many additional projects were added to the MURI, and in some cases, the experiments were carried out on different molecules than originally stated (e.g., laser cooling and trapping in a MOT).

### Summary of most important research outcomes

After the very first production of polar molecules in their absolute ground states, laser cooling and trapping (e.g., in a magneto-optical trap or MOT) and evaporative cooling was deemed close to impossible by many scientists prior to this MURI, as was reliable cooling into a complete molecular ground state. Nevertheless, all three were accomplished in our research. The first laser cooling, the first MOTs, the first ground state cooling, and the lowest external and internal temperatures were accomplished in those five years. These methods are, however, not universal to all molecules and not yet feasible for all labs, thus, in addition, we have developed simpler and more versatile techniques to reach the same goals, with less dramatic results but results that are potentially more important for future developments. This includes, most notably, the development of sophisticated buffer gas sources that allow for high-phase-space density ultracold samples of a wide variety of molecules. These techniques have now literally been distributed as the new gold standard to labs over the whole world. Also, molecule production with more specialized focus plays an important role, such as the (more involved) production of NaLi, the smallest polar molecule, which promises special properties, and the benchmark experiments on K and Rb in more modest laboratory settings that will be important for a wider group of scientists in the decades to come. This also includes the first studies of on-chip molecular ions.

Ultracold chemical reactions have been, theoretically and experimentally, been studied for the first time (and mostly exclusively) in this MURI group. A major part of present knowledge about controlling ultracold chemical reactions using electromagnetic fields, coherent methods, geometries, and other methods, in particular to study which channels for

which classes of molecules can be modified by different methods. The challenge we faced in this part of the project is the pure multitude of different possibilities, such that classification for certain chemical processes in this regime and how to understand and manipulate them was a major product of this MURI. This, in particular, includes also the new phenomenon of *sticking* that is relevant only to ultracold temperatures.

Suggestions for applications for ultracold molecules were deliberately kept somewhat vague in the proposal, since applications typically come out of special findings during the research and are often hard to predict. Thus, the outcome in this category of our MURI is a combination of the predicted, most notably a comprehensive study of the many-body physics of interacting dipoles in optical lattices theoretically and first major success to experimentally implement those. It is important to note that this touched (and often initialized) virtually all areas of recent interest in the theory of strongly interacting systems, such as magnetism, Majorana Fermions, many-body localization, to name just a few. In addition, there has been a multitude of less predictable outcomes: special quantum information processing schemes, uses of entanglement such a spin-squeezing for better quantum measurements, and the foundations of a method to distinguish molecules of opposite chirality (the method itself is not part of this MURI). Among the applications belong also the proposals and experimental implementation of improvement of detection methods.

## Itemized list of accomplishments:

### *Production, cooling, trapping, detection, and spectroscopy*

#### – Buffer gas sources

The Doyle group developed and demonstrated a novel approach to magnetic trapping of a wide variety of molecules using a two-stage buffer gas beam source in conjunction with optical loading into a magnetic trap. Our two-stage buffer-gas source produces molecules slow enough so that simply through the interaction of the molecules with the fringing fields of the trap, they can be slowed to trappable velocities. In order to load irreversibly, two lasers are used. The wavelengths are set to pump the molecules from low-field seeing states to high-field-seeking states (and back) at key points in the magnetic field. The molecules spontaneously emit photons as they are pumped, providing the irreversible steps. We fully modeled this approach for CaF and CaH and then implemented the procedure, verifying our quantitative model. We also did the same trapping with an atom, potassium, trapping large numbers. This opens the door to atom-molecule sympathetic cooling and study of collisions. We are now working on spectroscopy of SrOH, leading up to co-trapping of SrOH and Li (or K). In addition, buffer gas cooling methods have been used and developed by the Ye, DeMille, and Lewandowski groups.

#### – Trapping and cooling

Laser cooling in increasingly high dimensions has been accomplished by both the DeMille and Ye groups for SrF, TlF. Magnetic and magneto-optical trapping, often assisted by laser cooling in some dimension has been done for several species. This has been accomplished stepwise over the years, mostly one dimension at a

time, and increasingly combining the methods for best results. In addition, many of those efforts have been assisted by producing slow molecular beams with the help of the Doyle group. In general, with cooling methods, velocities of a few tens of m/s have been achieved. In addition, loading efficiency and densities have been approved, even to the extent that the Ye group has succeeded in evaporative cooling of OH.

- Stark deceleration

New Stark deceleration techniques, mostly developed in the Lewandowski group but also by Ye and DeMille, allowed strong increases in molecular density. Various types of molecules were cooled in analog-controlled voltage Stark deceleration. In particular, in the Lewandowski group, Stark decelerated CH molecules were used for studies of ultracold chemistry.

- Indirect cooling

First cooling atoms to ultracold temperatures and then associating them into molecules is one of the most versatile methods to produce ultracold molecules. This avenue has been followed by the groups of Eyler, Gould, and Stwalley (UConn expo) as well as the DeMille and Ye groups, often with theoretical support, notably by the Côté group and others. These efforts most importantly include magneto- and photoassociation of various molecules, such as KRb and RbCs. In addition, the Ketterle group has focused on producing LiNa this way, a molecule that is singled out by being the lightest bi-alkali molecule and thus having special properties such as chemical stability. Over the years, all these methods have been improved considerably, in particular with regard to volume, density, efficiency, selectivity of final states (e.g., production in to single hyperfine states). Since most of these molecules could not be studied in detail in the ultracold regime, extensive spectroscopic studies have been performed as well that should allow both, work based on those specification and easier spectroscopy of other molecules in the future. In addition, the UConn exp group has now started to study highly excited molecules, producing so-called trilobite states.

- Optical lattice ultracold polar molecules

Polar molecules (KRb) can now be produced in a 3D optical lattice in the Ye lab and are studied for quantum information processing.

### *Collisions and Chemistry*

- Calculations and specification

The Kotochigova group performed extensive calculations to characterize general ultracold collisions and support experimental groups. This included state-dependent potential surfaces, collision cross sections, potential energies and dipole moments, effects of electric and magnetic fields, new resonances, long-range coefficients, effects of confinement, and dynamic polarizabilities for a large number of different molecules. These calculations always are the first starting point for experiments, and her group is close to have the world monopolies on these calculations. In addition, the Côté group has found (potentially generalizable) properties

of the KRb system such as chemical stability of the various combinations of the two species and in particular the often overlooked but important influence of higher order poles such as quadrupoles.

- Geometry and anisotropy

The nature of anisotropic collisions and the use of direction-specific confinement was investigated by the Bohn and Ye groups. Reaction suppression and anisotropy was measured in different setups.

- Detection and spectroscopy

In all experimental groups concerned with chemistry (JILA, Temple, Yale) spectroscopic identification methods have been improved as well as detection methods such as direct absorption imaging as well as . At UConn, this has in particular included the novel trilobite molecules.

- Control using electric/magnetic fields

In the JILA groups, schemes were investigated to control the Stark shift of rotational states using lasers and electric fields. In addition, for OH, detailed studies were performed on the action of crossed electric and magnetic fields. Progress is being made with field-induced molecular collisions, state-selective molecular collisions, and quantum Zeno measurements of those.

- General collisions and universal properties

The Bohn group has studied cold collisions in complex systems (Er, Dy, OH), including the prediction and characterization of novel long-lived resonance states. In addition, they studied universal features among all dimers that can react at zero temperature. In addition, they found that ultracold collisions of alkali dimers may result in a novel “sticking” for milliseconds, with implications for trap stability etc., describable in the context of “quantum chaos.” The Ketterle has found general universality and particular exception to it in LiNa collisions. Other groups have studied the properties of specific systems such as molecule collisions with pnictogen (Nitrogen) and a molecule (NH), NH<sub>3</sub> and OH (Doyle), theory of the benchmark system H<sub>2</sub>+D (Côté), Rydberg dressing (Côté), Rb and Cs (DeMille), OH (Ye).

### *Quantum information and simulation.*

This thrust has been mostly moved forward by the theoretical groups and, in the second half of the project, by the JILA lab. The main research here consisted of

- Quantum optics and quantum information with ultracold polar molecules

In the Côté and Yelin groups, we proposed the use of Rydberg-mediated polar molecule interactions, methods to produce cluster states using dipolar interactions, and compared the use of polar molecules and ions for single photon nonlinearities, realistic quantum information processing schemes with molecular ions and anisotropic effects in optical lattices, cluster states in optical lattices, and a hybrid atom-molecule platform for quantum computing.

In addition, the Yelin group showed how ubiquitous cooperative effects (i.e., superradiance) is in ultracold molecular systems, in particular, their vibrational and lower-frequency transitions. and proceeded to study entanglement and spin squeezing in these systems, which can be considerable, both in ideal and in realistic systems. Collaborations with experiments have started recently.

- Dipolar many-body physics:

Most of the theoretical work done in the Demler group concerns quantum simulations of many-body systems with (dipolar) interactions in single and multiple layers, such as exotic spin textures in magnetic superfluids, exotic paired states and Majorana fermions as well as Shiba states in fermionic superfluids, few-body bound states, impurities to find attractive polarons and manifestations of Anderson’s orthogonality catastrophe, realizations of topological states using polar molecules on a lattice, studied the effect of dipole-dipole collisions on the transport properties in 2D many-body localization, a new class of unconventional critical phenomena characterized by singularities only in dynamical quantities, ways to experimentally probe such states and find new phases and resonances. This includes (i) systems of strongly interacting dipoles to study many-body localized phases, (ii) motion of an impurity particle injected with finite velocity into an interacting 1D quantum gas using large-scale numerical simulations, resulting in so-called *quantum flutter*; (iii) the dynamics of magnetic correlations in the half-filled fermionic Hubbard model following a fast ramp of repulsive interaction, leading to different dynamical behavior.

The Yelin group has modeled quantum phases of quadrupolar particles, such as some homonuclear molecules, and are now studying quasi-1D and 2D many-body effects of particles with resonances due to in-plane dipole moments, with plans to investigate, in particular, the transition  $1D \rightarrow 2D$ , short range  $\rightarrow$  long range interaction, and frustration.

- Experimental efforts with molecules in optical lattices

The Ye/Jin group has realized a molecular spin lattice using dipolar exchange interactions in a 3D optical lattice, using it to study quantum magnetism and to compare to theories in this context. Encoding spin in rotational states, this led to observing spin exchanges of ultracold polar KRb molecules that are confined in a deep three-dimensional optical lattice. These observations were made in a regime where the lattice filling is dilute. Current experimental efforts are focused on increasing the filling fraction with plans to enable the study of richer physics, such as transport of excitations in an out of equilibrium long-range interacting system.

### *Molecular Ions*

The Chuang group at MIT built an integrated on-chip superconducting ion trap for molecular ions. In particular, over the years, redesigned traps improved the major problem of charging dielectrics due to laser ablation in traps significantly thus demonstrating the trap. In addition, they performed detailed calculations for interfacing polar molecules with circuit QED systems and modeled photodissociation of molecular



ions, particularly  $\text{BaCl}^+$  (with some help from DeMille). After another redesign, a new cryostat allows to increase microwave power to the superconducting trap chip, and a new chip design holds the ion  $190\text{ }\mu\text{m}$  above the surface and thus increases the effective field strength.  $\text{BaCl}^+$  serves as example on how we calculated electronic properties of dipolar ionic molecules, with focus on sympathetic cooling. A general method for rotational microwave spectroscopy and control of polar molecular ions via direct microwave addressing is considered, which would allow to make use of spatially varying AC Stark shifts, several unrelated factors led the group to abandon further development of the project.

A new project in the UConn experimental group has been started: they worked on the possibility of photoassociative ionization to form  $\text{Rb}_3^+$ . This work will be continued with different support. In addition, in the Kotochigova group, electronic properties of molecular ions with a special view at sympathetic cooling have been performed.

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